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# The Final Technical Report of AFOSR Grant #447288, Aug. 31, 1995

## Research Title:

**"Nonlinear photon localization for high intensity laser protection systems for photodetectors and the eyes"**

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The goal of research was to investigate new phenomena and to develop new optical devices using random layered and bulk materials for laser and industrial applications. The following summarizes the research accomplished in: (1) light propagation and (2) lasing in random media.

### 1. Light propagation in random layered and bulk materials

The goal of this research was to design and fabricate optical power limiters using multilayered nonlinear materials and studying their transmission and pulse dispersion properties. Four areas have been investigated.

#### (1). Nonlinear photon localization for high-intensity laser protection system.

We have shown that a random multilayer of nonlinear materials can be designed to allow the ambient light to pass through but reflect the high-intensity laser beam using the principles of photon localization and nonlinear optics. This random nonlinear multilayer system may be an ideal active protective optical radiation device covering an extremely broad spectrum, turning on and off with the intensity of the optical radiation.

The underlying physical principles for the operation of the novel protective eyeglasses are based on photon localization and nonlinear optics. Photon localization of the random one-dimensional system gives rise to the characteristic of broad bandwidth of high reflection. In a one-dimensional system, such as the multilayer dielectric system, light cannot pass through but will be totally reflected over a broad spectrum if the phase shift (or the optical thickness) for each layer fluctuates randomly. Total reflection will occur even with an infinitesimally small amount of randomness in phase shift if the number of layers is sufficiently large. The transmission of the low-intensity light is determined by the linear refractive indices of the multilayer system, which can be designed to transmit the ambient light totally. In order to reduce the transmission of the multilayer

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system as the radiation intensity increases, the system is made of alternating high and low nonlinear materials, and the thickness of each layer is randomized. The refractive indices of each layer can then be changed and regulated by the radiation intensity by means of the induced nonlinear refractive index. These induced refractive indices change the interface reflectance and randomize the phase shift of each layer, creating a random one-dimensional system. This induced random one-dimensional system regulates the reflection of the radiation and actively depends on the radiation intensity. The higher the radiation intensity, the higher is the change in reflective indices, which results in higher reflection for the higher intensity light.

The reflectance of a multilayer system can be computed numerically by Rouard's method and is determined by the reflectance at the interface  $k$ ,  $r_k$ , and the phase shift at layer  $k$ ,  $\delta_k$ , which are given by

$$r_k = (n_k \cos \theta_k - n_{k+1} \cos \theta_{k+1}) / (n_k \cos \theta_k + n_{k+1} \cos \theta_{k+1}), \quad (1)$$

$$\delta_k = 2\pi n_k d_k / (\lambda \cos \theta_k), \quad (2)$$

where  $n_k$  is the refractive index,  $\theta_k$  is the angle of incidence,  $d_k$  is the thickness of the layer, subscript  $k$  refers to layer  $k$ , and  $\lambda$  is the wavelength of the light.

The refractive index of the  $k$ th layer if written as

$$n_k = n_{lk} + n_{nlk}, \quad (3)$$

where  $n_l$  and  $n_{nl}$  are the linear and nonlinear refractive-index components, respectively. The nonlinear index is proportional to the light intensity,

$$n_{nl} = n_2 E^2, \quad (4)$$

where  $n_2$  is the nonlinear index coefficient. The value of  $n_2$  for materials varies from  $10^{-4}$  to  $10^{-12}$  esu depending on the response time of the material. The refractive index of the material for low-intensity light is equal to  $n_l$  since  $n_{nl}$  is negligibly small. The ambient light transmission in the multilayer system is determined by the linear component of the refractive index. A well-known example of a multilayer system that transmits the ambient light is an anti-reflecting coating.

The reflectivity or transmittivity of the random system depends on three parameters of the system: the layer interface reflectance  $r$ , the fluctuation of layer thickness  $\sigma_s$  (measured in phase shift), and the number of the layers  $n$ .

At higher intensity the mirrors become to totally reflective due to nonlinear-index becoming active in random layers. The nonlinear multilayer system is its active response to the intensity of the radiation: the higher the radiation intensity, the higher the rejection. The higher radiation intensity induces a larger refractive index, which results in higher interface reflectance  $r_k$  and larger fluctuation of phase shift  $\delta_k$ . These changes increase the reflectance. The incident light completely reflects when the mean layer thickness is

from 300 to 1300 nm,  $n_{nl}$  is  $0.8n_l$ . The rejection of light above critical intensity level also depends on the number of layers in addition to the change in the refractive indices. When the number of layers is 2000, the system totally reflects the light over a broad bandwidth.

The major advantage of the nonlinear mirror designed with a random layer thickness over that with a predetermined layer thickness, as in the conventional broadband mirror, is that the random system will generate the broadest bandwidth of reflection.. This extreme broadband reflection is necessary in order to protect the detector over all possible laser radiation spectra.

(2). Numerical simulation in term of interface reflection, fluctuation of layer thickness, and number of layers on wave propagation for finite random multilayered media.

Photon propagation in nonabsorbing disorder multilayer systems has been studied as a function of different system variables. For systems with ensemble averaging, the transmission of a wave through a system of random multilayers decreases exponentially and then decreases more slowly as the number of layers increases. The random system has a maximum reflectivity when the fluctuation of layer thickness is around  $\sigma_\delta \cong 0.4\pi$ . For systems with small fluctuations, there is a range in which the reflectivity increases linearly with  $\sigma_\delta$ . If the gradient of this linear increase is greater than the critical gradient of 2.7, then the system can become opaque for same larger  $\sigma_\delta$ , with the interface reflectance and the number of layers remaining the same. For systems without ensemble averaging, a large fluctuation of reflectivity with sharp spikes as randomness increases is expected and shows a sudden surge in reflectivity as the randomness increases from zero. The features presented above and photon localization are shown to be easily observed experimentally in random multilayered system.

We present another system where the optical thickness of each layer remains constant at 250nm, but the refractive indices of the layers are random, that is, reflectance  $r$  at each interface of the layers is fluctuating. The reflectivity improves as the number of layers increases. The reflectivity improves significantly when the reflective indices of the layer fluctuate over a larger range. A extremely broad bandwidth multilayered mirror may be fabricated by randomizing the optical thicknesses of the layers. The bandwidth of reflectivity improves with the larger number of layers and lager fluctuation of optical thicknesses and refractive indices of the layers.

(3). Random-thickness-multilayered nonlinear system

A system of 30 layer random thickness composite was fabricated consisting of alternating layers of  $\text{TiO}_2$  and  $\text{SiO}_2$  where  $\text{TiO}_2$  is known to have a much higher  $n_2$  index than  $\text{SiO}_2$ . The change in the amount of intensity dependent dispersion of an ultrashort pulse after transmission through the random-thickness mirror was observed. The increase in dispersion due to increase in intensity has been attributed to the change in index of refraction,  $n = n_l + n_{nl}$  where  $n_l$  is the linear index and  $n_{nl}$  is intensity dependent nonlinear index.

Additional properties of such composites are being evaluated. The change in the transmission spectrum as function of angle is investigated.

The random stratified composites were observed to have an interesting property of random phase dispersion for any given wavelength. A careful manipulation of the thickness parameters can yield any given phase dispersion for any given wavelength region. This in turn gives a control over the group velocity dispersion and group delay dispersion of pulses. Thus with these composites artificial materials/systems can be fabricated to tailor the phase dispersion properties of a material to any given specifications. Such system have applications in the control and compensation of the chirp and pulse dispersion in ultrashort pulse generation system such as short pulse lasers. The first, second and third order phase dispersion  $\partial^2\theta/\partial\omega^2$ ,  $\partial^3\theta/\partial\omega^3$  characteristics of the random-thickness mirror sample is calculated.

#### (4). Nonlinear optical interaction in scattering media

Three nonlinear effects in random media were investigated: i) The intensity dependent nonlinear index,  $n_2$  in different scattering media, ii) Raman gain in nonlinear scattering media and iii) backscattering in nonlinear scattering media.

i). The nonlinear coefficient of  $\text{TiO}_2$  in  $\text{CS}_2$  were studied using the Z-scan technique. The self-focusing effect in  $\text{CS}_2$  was measured. When the nonlinear material  $\text{TiO}_2$  was added to the  $\text{CS}_2$ , the transmission T (peak to valley) changed was investigated. The difference is attributed to the net change of  $n_2$  of the system. The  $n_2$  is observed to have been altered in the  $\text{TiO}_2/\text{CS}_2$  solution where a typical value of  $T_{\text{CS}_2 + \text{TiO}_2} / T_{\text{CS}_2}$  is 1.32. There appears to be a gain of 30% in  $n_2$ .

ii). Raman gain of nonlinear random media was measured using OMAIII system and BOXCAR integrator method using a laser pulse and Stimulated Raman Scattering pulse from ethanol. The Raman gain in a solution of  $\text{TiO}_2$  in ethanol and  $\text{TiO}_2$  in water (O.D. = 0.8) was measured. No significant gain was observed in the  $\text{TiO}_2/\text{ethanol}$  solution over  $\text{TiO}_2/\text{water}$  solution.

iii). The backscattered coherent peak in two-photon-absorbing dye solution was observed for a solution of polystyrene balls in water and different concentrations of a two-photon absorbing dye, Malachite Green. The change in the backscattered angular profiles for different intensities of laser pulse was investigated. The reduction in weak localization peak via two-photon-absorption was observed using a CCD camera.

## 2. Lasing action of laser material in scattering particle suspension media

Three areas were investigated.

(i). Laser action of dye in highly scattering media is observed. The threshold for stimulated emission is dependent on the concentration of both laser dye and scattering particles. A  $2.5 \times 10^{-2}$  M dye concentration, the lasing threshold was found to be reduced

by more than two order of magnitude in comparison with conventional lasers when the density of scattering particles was  $2.5 \times 10^{12} \text{ cm}^{-3}$ .

(ii). Experimental measurements were carried out on both classes of disordered media, i.e. discretely disordered and continuously disordered media, to explore their nonlinear optical characteristics. Laser action was observed from both classes of disordered media. Animal tissues were used as the continuously disordered structure while dielectric particles in active dye solution host were used as the discretely disordered media. Laser action was also observed from densely packed dielectric powders in dye hosts. Data on lasing characteristics such as shortening on temporal and spectral emission profiles and input intensity thresholds was collected and published.

(iii). Spectral and temporal behaviors of laser action of dye in suspension of different size of particles have been measured. The experimental data of lasing threshold depend on particle size are agree with the calculated results from Mie scattering theory.

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